

Light Absorbing Particle (LAP) Measurements in the Lower Stratosphere



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Photo courtesy of R. Shetter

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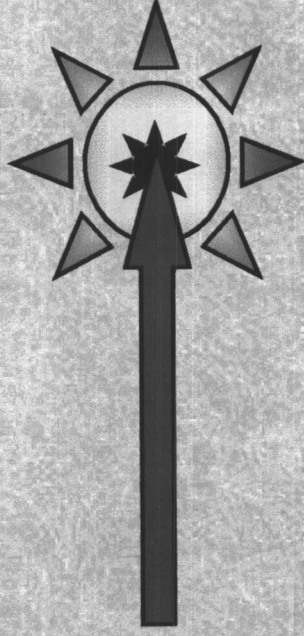
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David Fahey and Ru-Shan Gao

SOLVE II Science Team and Flight Crew

Single Particle Soot Photometer

SP-2*



Particle size using light scattering
range limited by electronics

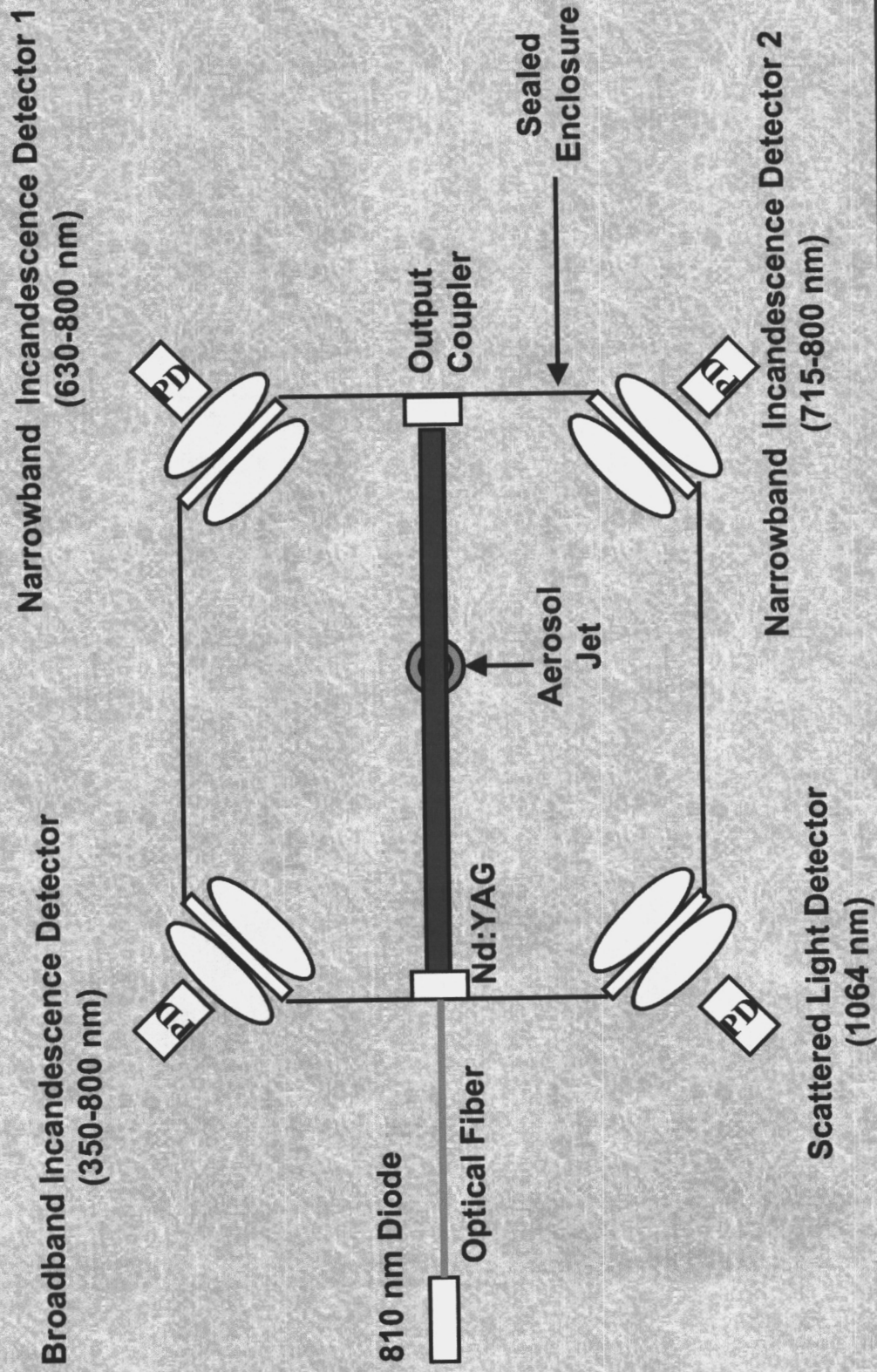
0.2 - 1.0 μm ($\pm 30\%$)

Particle mass by laser induced incandescence
*accuracy limited by calibration
and uncertainties in response to mixtures.*

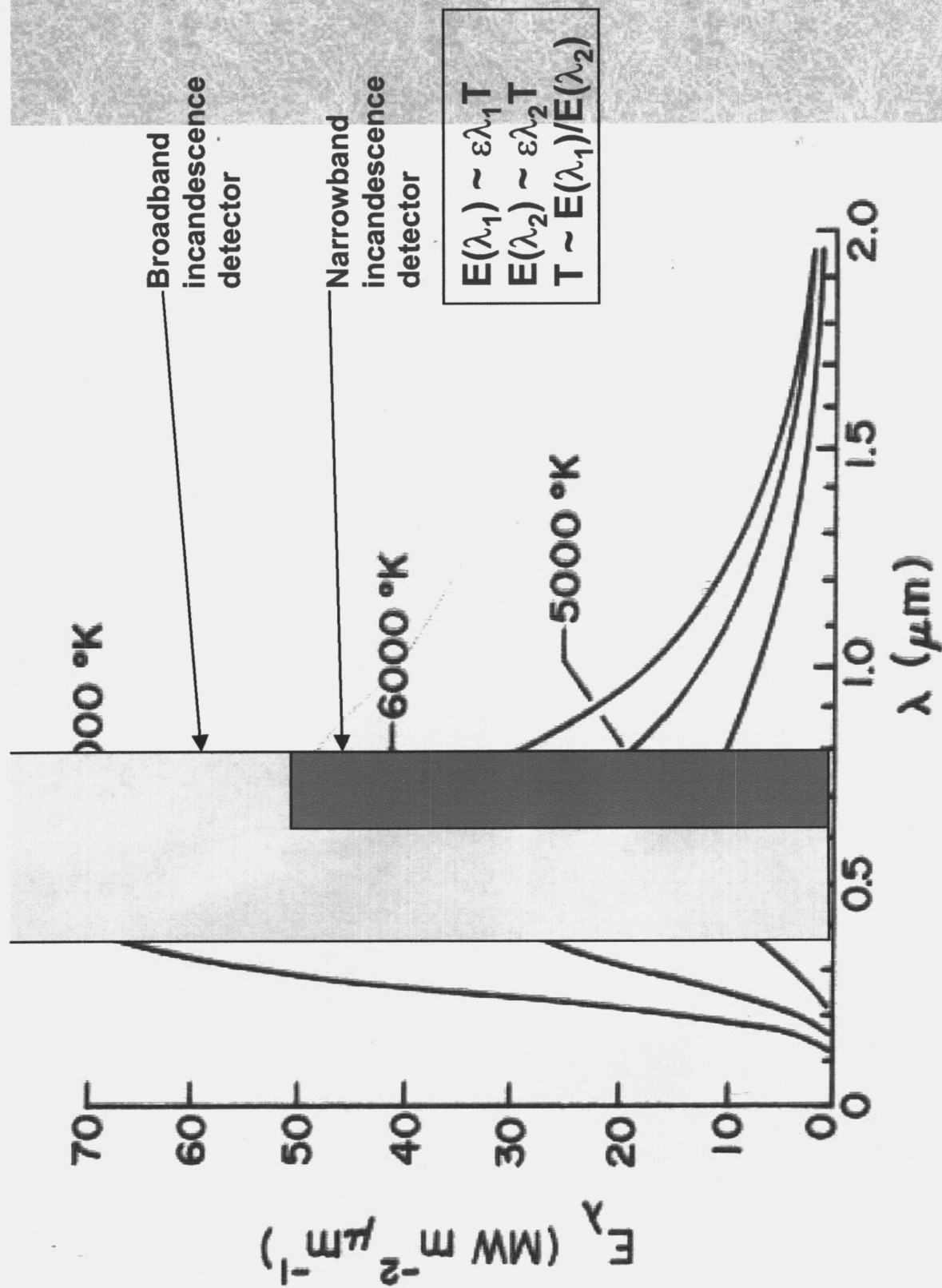
.02 pg - ? ($\pm 50\%$)

Estimate of particle composition from incandescence temperature
black carbon and other light absorbing particles (LAP)

* Developed by Droplet Measurement Technologies under ONR SBIR program



Aerosols pass through the center of focus of the 1064 nm laser beam that has a power density of approximately 1Mw. The scattered light of each particle is collected by the detector with a filter at the wavelength of the laser. Particles that absorb light will heat and emit photons at the wavelength proportional to the temperature. Three detectors collect this light over different wavelength bands.



The ratio of light measured in the different wavelength bands is independent of emissivity and from this ratio is derived the temperature of incandescence. This incandescence temperature is an indicator of the particle composition, e.g. black carbon incandesces at approximately 4200 K while iron has a temperature of incandescence of approximately 2850 K.

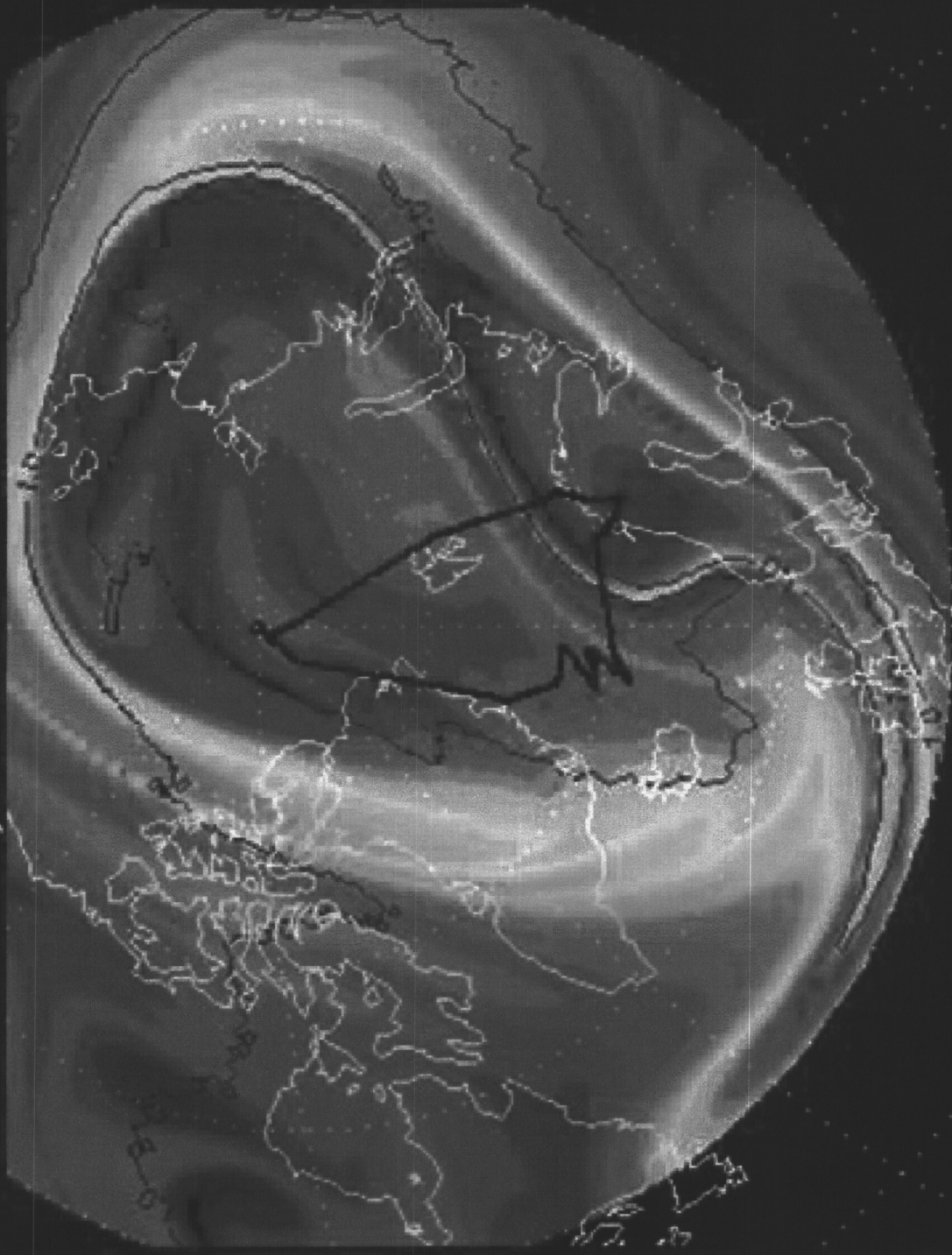


The SP2 was deployed on the NASA DC-8 during the SOLVE II field campaign from January 15 - February 6, 2003. The SP-2 made measurements during the last 6 flights of this project.

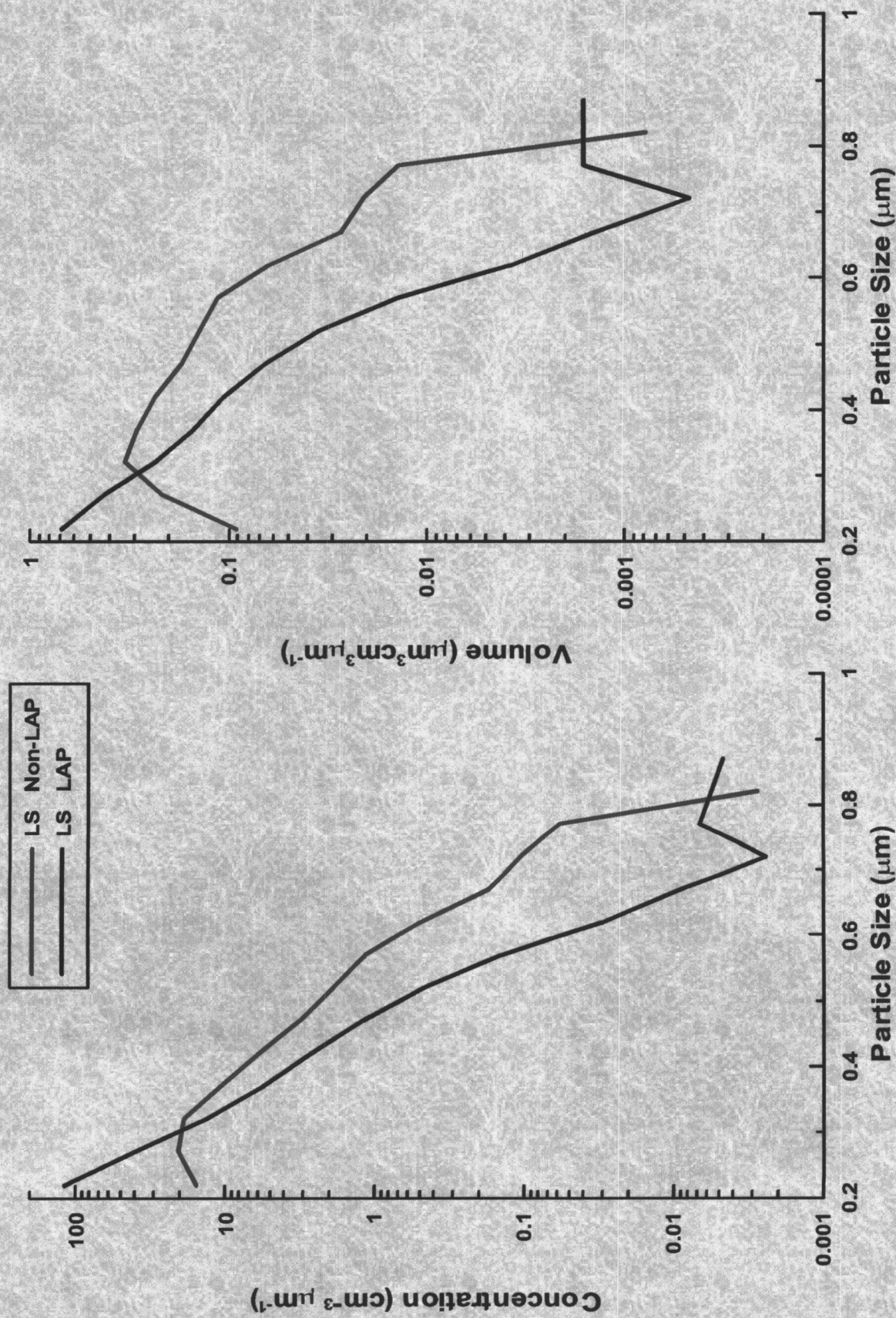
The SP2 was mounted in a rack, shared with the NASA Langley aerosol group that was measuring total and non-volatile CN and size distributions with an PCASP, FSSP-300 and CAPS probe. The CN counters and SP-2 sampled from the same. forward facing inlet.



4day RDF for 48DK Feb. 2 2003



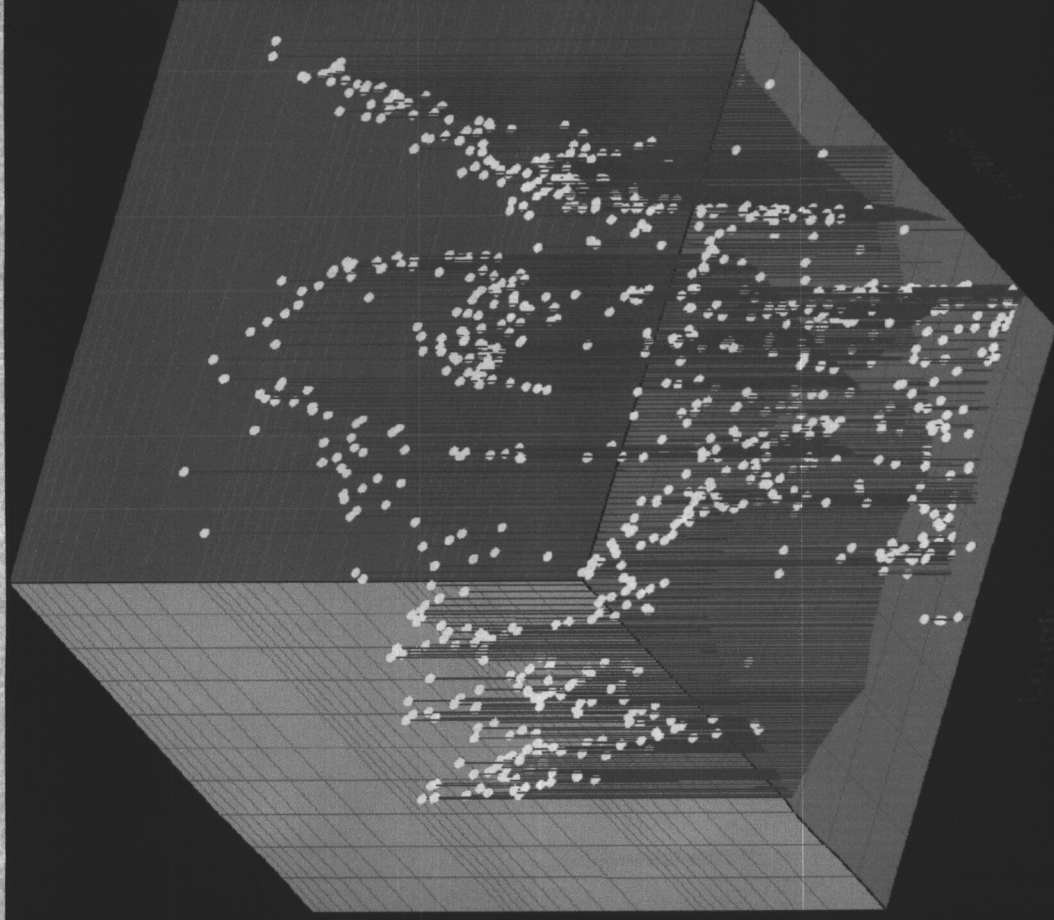
The majority of the flight patterns took the aircraft northwest from Kiruna, Sweden, where the aircraft was based, into the Arctic Vortex. The aircraft flew as far west as the west coast of Greenland and as far north as the north pole. The flight altitude was normally between 9 and 12 Km.



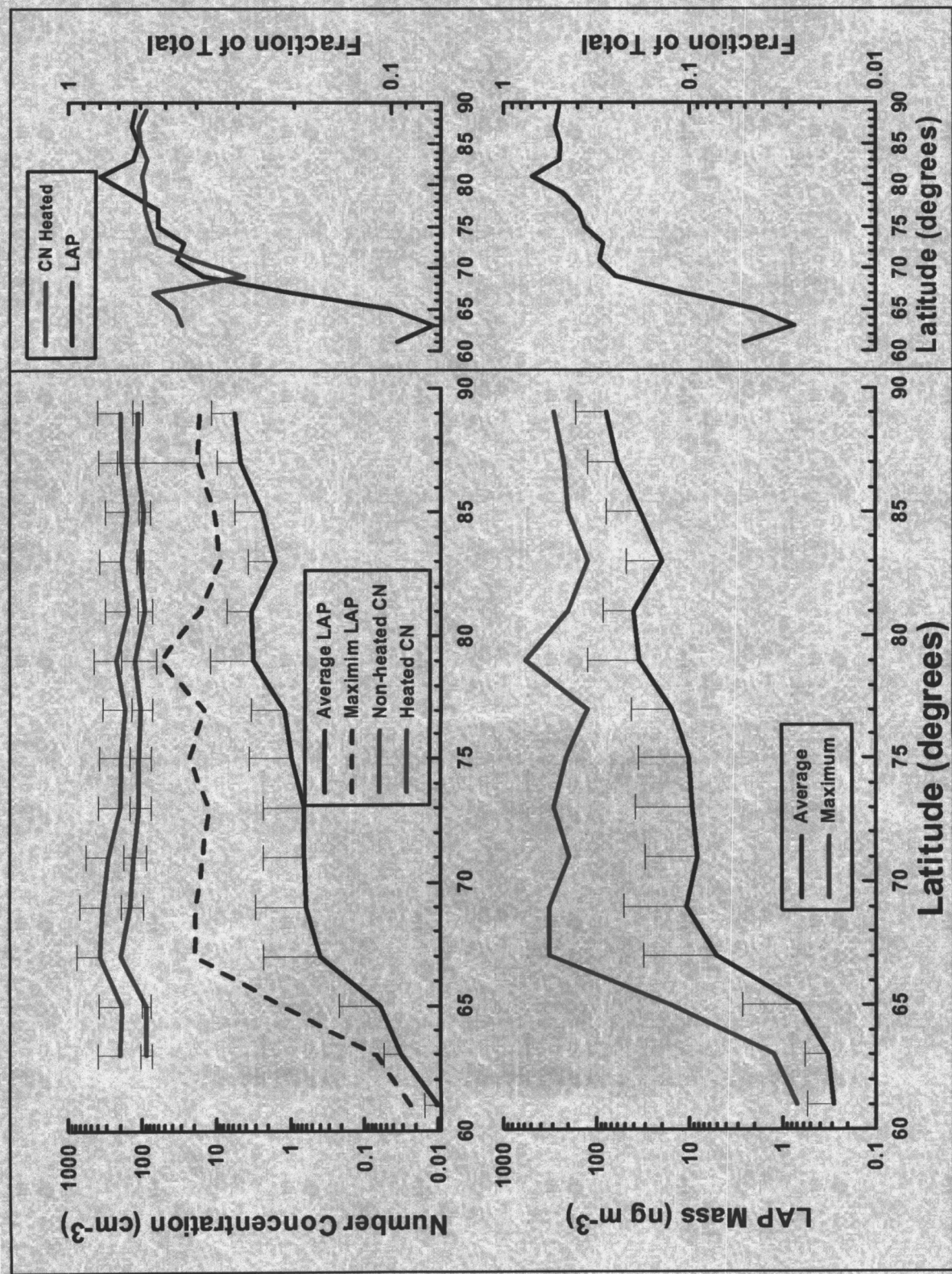
These size distributions of number and volume concentration as a function of size are averages over all six flights but only in the stratosphere, where the stratosphere is defined by ozone values greater than 100 ppb. Note the much higher concentrations of absorbing particles less than 0.4 μm .

Lower
Stratos

1 min. Averages



This 3D representation shows the mass of light absorbing particles as a function of location. There is a great deal of variability but in general, mass increases to the west and north, with a range from 1 to 600 ng m⁻³.



The SP2 number and mass concentrations were averaged in 2° intervals of latitude for stratospheric altitudes. Also averaged were the concentrations from the heated and non-heated CN counter. There is a positive trend in LAP concentrations, with a fair degree of variability. The fraction of the total concentration that are LAP particles (right panels) also increase with latitude. The LAP fraction is consistent with the non-volatile CN fraction that was measured.

Research Study	Number Concentration (cm ⁻³)*	Mass Concentration (ng m ⁻³)*	Number Fraction	Mass Fraction
Pueschel et al. (1992; 1997)	?	0.03 – 0.18 <X> = 0.07	?	.0001
Blake and Kato (1995)	0.07	0.02 – 0.8 <X> = 0.25	0.02	0.03
Strawa et al. (1999) ^{***}	0.07-0.6 <X> = 0.2	0.4 – 6.8 <X> = 2.2	0.01	?
Current Study	0.01 – 30 <X> = 1.8	0.3 – 600 <X> = 23.6	0.07 – 0.8 0.3-0.6 (CN) ^{**}	.03 – 0.7

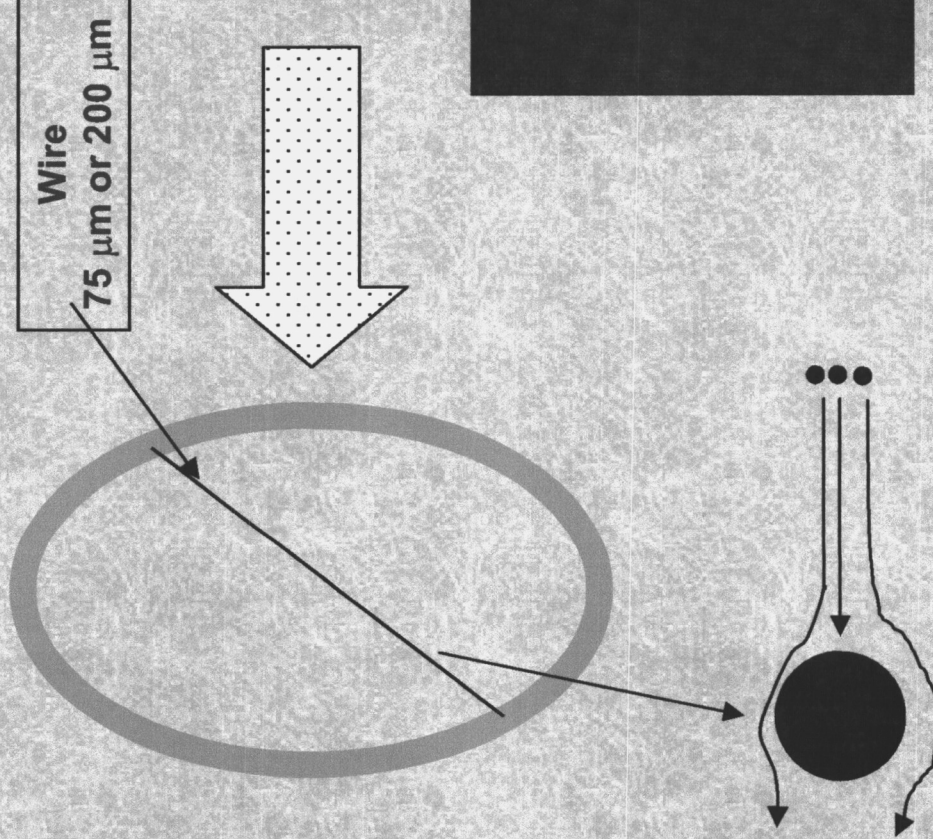
*** Adjusted to 230 mb, 210K**

**** Heated to Unheated**

***** 18 Km**

This table compares the current measurements with previous measurements in the same latitude and altitude band.

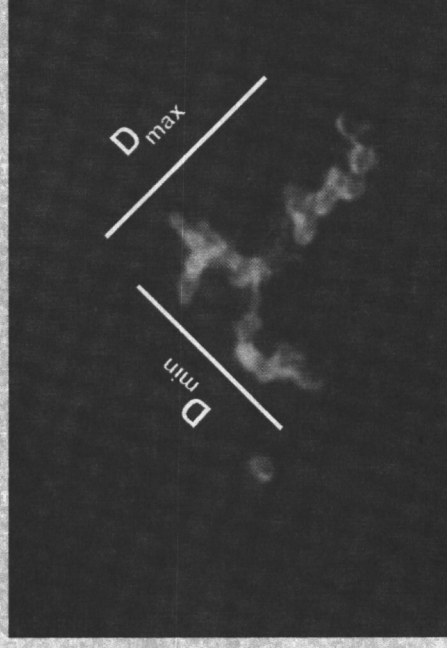
Wire Impactor



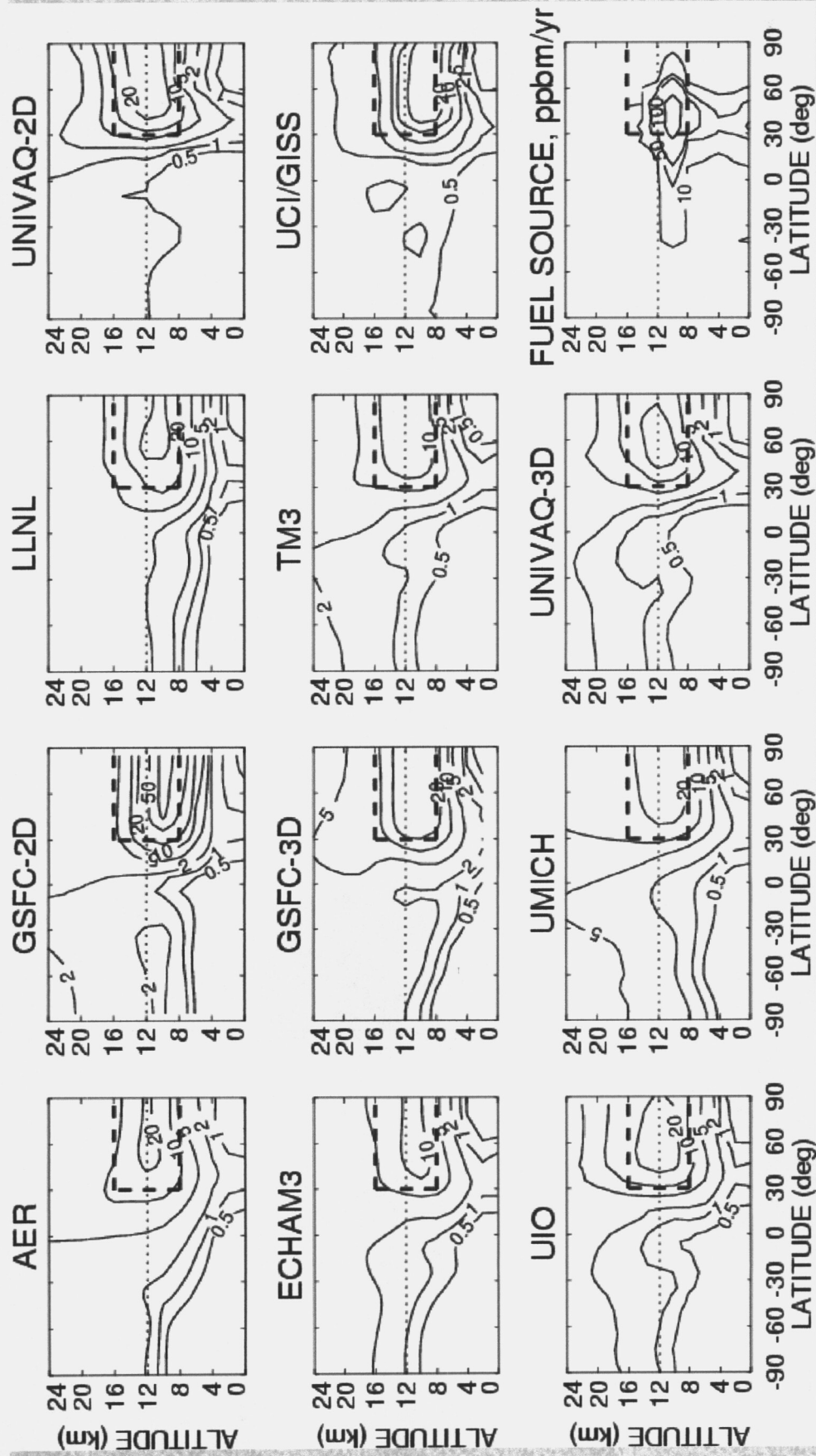
$$D = \frac{D_{\min} + D_{\max}}{2}$$

$$V = \pi D^3/6$$

$$\text{Error} = \pm 1000\%$$

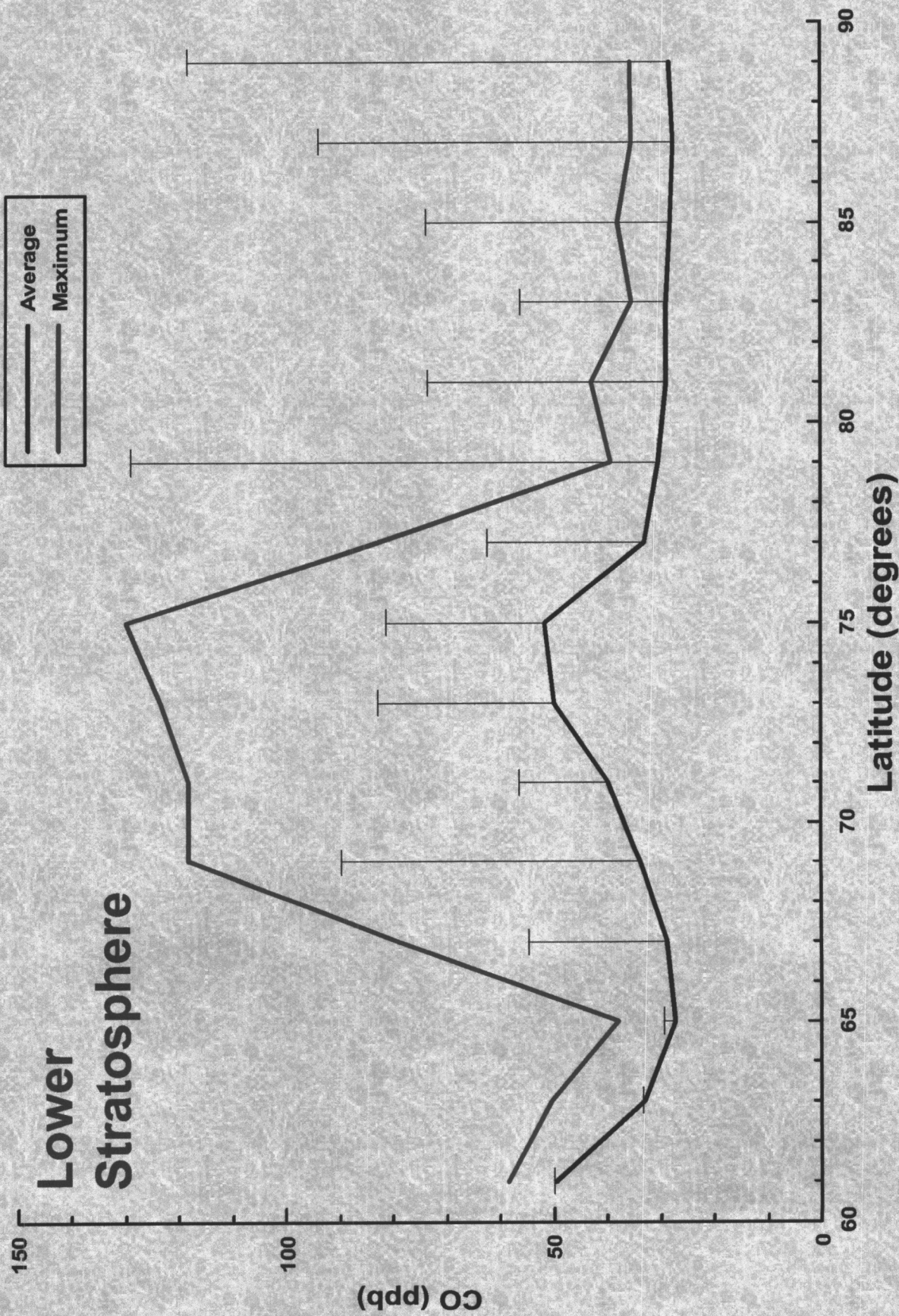


Previous measurements used wire impactors to collect particles and electron microscopy to estimate black carbon mass. There are huge uncertainties associated with this technique.

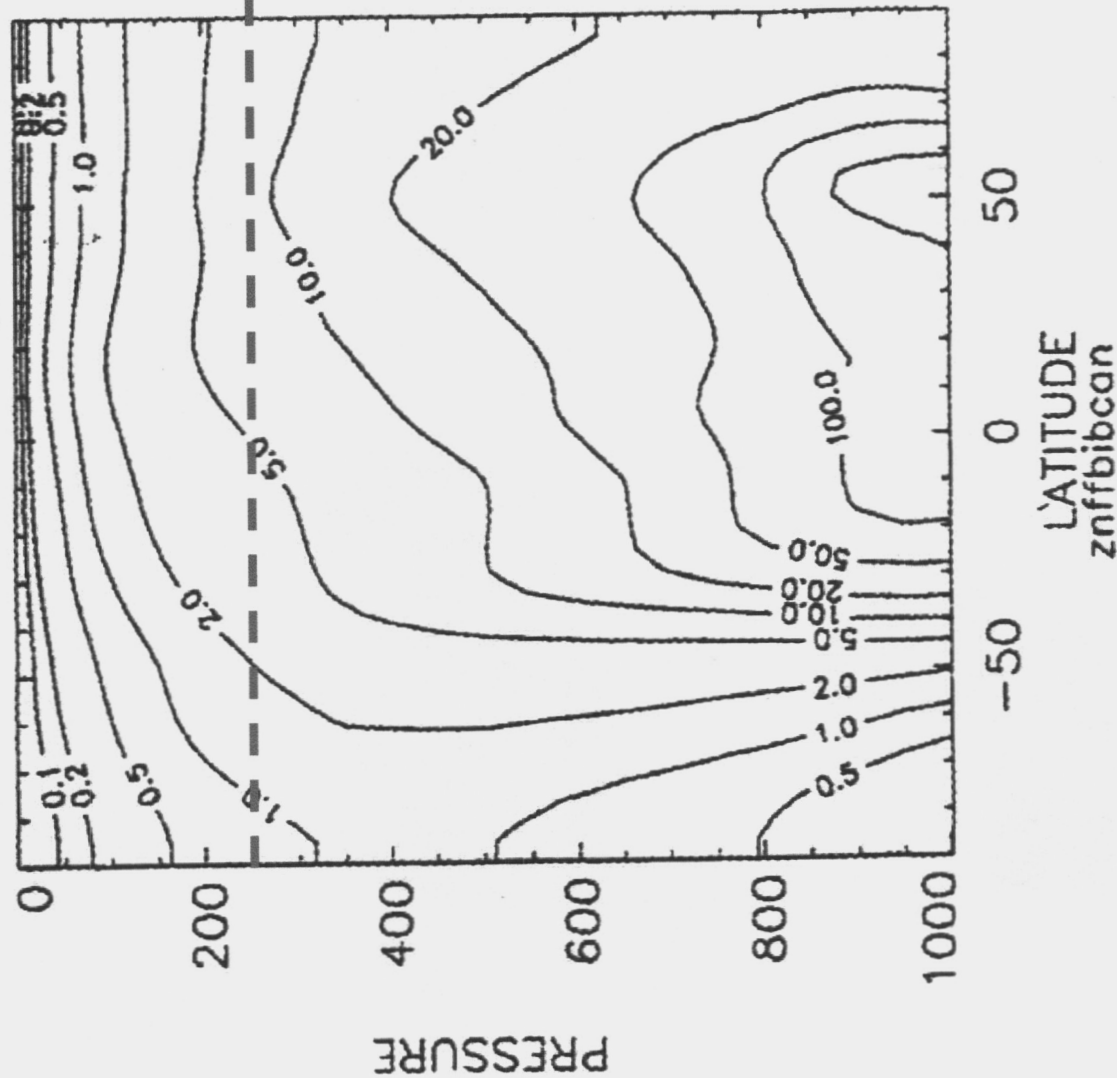


latitude	55	55	65	50	80	55	55	65	50	58±9
ng/m3	0.39	1.80	1.10	0.54	0.19	0.69	0.30	0.45	0.57	0.6±0.5

These model results show a tracer experiment for simulating aircraft emissions and their global transport. By multiplying the tracer value by an emission index of 0.04 kg/kg for black carbon, the equivalent carbon mass can be estimated. The table shows results from the 11 models, where the latitude is where the maximum tracer material is found. In red are the average. The point here is that the maximum values are about 20 times smaller than the average SP2 values.



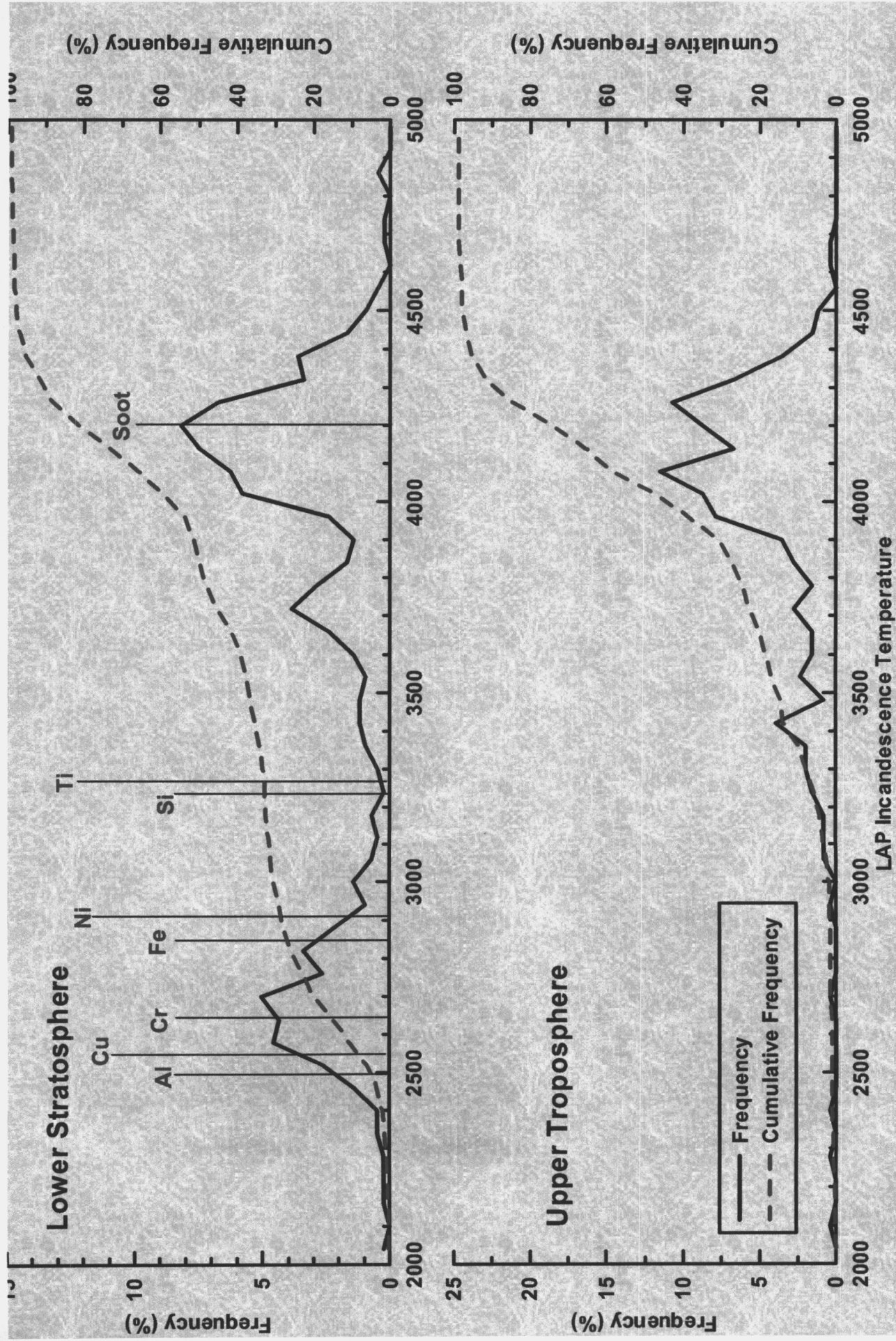
The CO measurements, averaged in the same latitude bands, indicate large variability in CO above the background stratospheric value of approximately 30 ppb. This suggests the transport of tropospheric air into the arctic vortex. This implies that a large fraction of the absorbing particles are coming from sources other than aircraft and from the troposphere, perhaps biomass burning or urban emissions.



latitude	ng/m ³
60-70	3
70-80	16
80-90	46

Figure 13b. Same as Figure 13a, but for concentrations of black carbon (in ng C/m³).

Global 3-D models of carbonaceous aerosols (e.g., Liousse, 1996) show black carbon values in the Arctic regions that are comparable to those measured with the SP-2.



The frequency distribution of incandescence temperature shows an interesting contrast between tropospheric and stratospheric regions. Only LAP representative of black carbon is seen in the troposphere, while particles with lower incandescence temperatures are found in the stratosphere. Several different types of metals, found both in meteorites and in aircraft emissions, would have this type of signature.

Questions

- 1.) Can the BC mass discrepancies of 10-100 be explained by differences in measurement techniques?
- 2.) Can increases in aircraft emissions over the past 10 years explain differences between the measurements in 1992 and 2003?
- 3.) Do the current measurements suggest that tropospheric sources of BC are more important than aircraft? Can urban emissions be playing a significant role?
- 4.) How do we explain the large fraction of metals in the lower stratosphere that are not measured in the upper troposphere? Meteorites? Washout in UT?
- 5.) How efficient is wet deposition for removing BC?